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DERIVATION OF BIOASSAY SELECTION CRITERIA FOR TRITIUM-- FROM CONSIDERATIONS OF THE RELATIVE RADIOTOXICITY OF VARIOUS COMPOUNDS AND EXPOSURE EXPERIENCE IN VARIOUS FACILITIES*

by

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A review** of the literature on the acute and chronic effects of tritium at the human, animal, tissue, cellular, and sub-cellular levels is now beginning to reveal a fairly consistent pattern. It is now possible to assign ranges to the effectiveness of various tritiated compounds, HT or T_2 gas, and tritiated DNA precursors, relative to HTO in producing the various biological endpoints investigated. A tentative selection of "radiotoxicities," relative to HTO has been made as follows: 1 for non-DNA precursors other than tritium gas, 10 for tritiated organic compounds that are precursors to nucleotides that can be incorporated into DNA, and 1,000 for tritium gas assured to be free of HTO or T_2O. With this conservative selection (on the safe side) of relative radiotoxicity, and experience (summarized in this paper) with the actual human intakes of tritium from various types of processes, suggested criteria are proposed for judging the need for bioassay monitoring of personnel. (These suggested criteria are taken from draft material under preparation by the author, are presented for purposes of discussion,* and do not represent official guidance of either

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the Nuclear Regulatory Commission, nor the position of the professional groups presently engaged in developing tritium bioassay guidance.[6,7])

Conclusions

1. In order to obtain more precise ordering of relative radiotoxicities, data are needed on: the intake of various organic compounds of tritium after inhalation, ingestion, and injection by animals and man; fractional concentrations and turnover rates in organs, tissues, and sub-cellular components; the biological effectiveness of such exposures compared to HTO exposures of the same activity; and relative doses in rads averaged over specified regions of interest. Too often, literature information on dosage or relative biological effect is ambiguous in regard to the specific regions over which dosage is calculated; or for sub-cellular components, whether dose is averaged over all cells of a type or only those that have absorbed tritium and have been exposed to some degree of damage by beta radiation. An attempt should be made to provide a set of controls exposed to HTO, and a set exposed to H_2O, under the same experimental conditions, for all experiments on toxicology of tritium compounds.

2. Despite uncertainties in toxicologic experiments, relative radiotoxicities (to HTO) of 1000^{-1}:1 r HT or T_2, and 10:1 for tritiated nucleotide precursors, appear from the literature to be safe assumptions for purposes of planning tritium bioassay programs.

3. Fractional amounts of tritium in various processes that have been taken into workers are less than 10^{-5} (average) under the poorest routine working conditions, and are usually less than 10^{-6}. A single incident has been recorded in an academic institution where a small spill resulted in an almost 10^{-2} intake of the material spilled, but the annual average exposures of employees in this academic institution resulted in an average intake of less than 10^{-6} of the material in processes or experiments. These orders of magnitude of intake are similar to the limits on fractional intake from accidents, as reported by Franke, Hermann, and Hunzinger ~~(9)~~ (9,102) More data on probabilities, or fractions, of intake are needed for a firmer assessment of exposure potential in working with all types of tritium compounds in various types of processes.

graduate student

4. The proposed activity levels in process (Table 2) above which bioassay for tritium is required are sufficiently low that (with bi-weekly or monthly sampling) dose commitments exceeding 300 mrem per quarter (averaged over body water) are not likely to go undetected. Also, these criteria would not generally be considered unduly burdensome by most health physicists experienced in tritium monitoring.

DERIVATION OF CRITERIA FOR DETERMINING THE NEED FOR A TRITIUM BIOASSAY PROGRAM

Introduction

The need for tritium bioassay of employees depends in general on many interrelated factors such as the nature and amount of tritium in process at any time, the types of processing, the extent of other safeguards or facilities provided to prevent occupational exposures, and the probabilities that significant quantities of tritium may be inhaled or ingested with the particular materials, procedures, and facilities involved.[1] Thus, the choice of an optimum combination of safeguards, including bioassay, is a complex problem and should be carried out by the laboratory supervisor in consultation with a professional health physicist experienced in tritium operations. However, experience[2-5] with the processing and use of tritium in various forms now allows the development of reasonable guidelines for determining the need for bioassay sampling and analysis under certain defined circumstances of operation.

Two groups have been active during the past year in developing guidelines for establishing tritium bioassay standards: the Health Physics Society Subcommittee (of the Standards Committee) on Internal Dosimetry Standards for Tritium[6]; and a subcommittee established by the informal Radiation Safety Officers' Conferences.[7] This talk--so far representing only my own views of the situation--is a summary of some information and ideas assembled in preparation for participation in the first of these efforts; a document with a more detailed review of radiobiological and applied health physics data may be obtained on request. I am handing out a questionnaire to obtain a broader range of data and opinions on questions related

to the development of standards of radiobioassay of tritium. Please fill out this questionnaire and send it to me to help in developing more reasonable guidelines consistent with your own experience.

General Internal Dose Levels Requiring Monitoring

Advisory and regulatory bodies have taken very similar positions regarding the need for monitoring radiation workers potentially exposed to external penetrating radiation. These can be summarized, for the sake of this standard, as the recommendation or requirement that appropriate personnel dosimeters be worn if there is a likelihood of receiving within a calendar quarter a dose equivalent (DE) in excess of 25% of the quarterly maximum permissible DE. For a whole body exposure, as generally assumed in the case of an exposure to tritium gas* or tritiated water, this number is 25% of 5/4 rem, or 0.3 rem per quarter.

Thus, this DE (0.3 rem) is believed to be a reasonable guide number on which to base the need for a bioassay program. About 3 mCi of tritiated water assimilated into the body tissue of a standard man (or 1.8 mCi using an RBE of 1.7 rather than 1)[12,14,39,84] will give this DE. This is consistent for our purposes with the Derived Investigation Level of 1.5 mCi intake, which would deliver 0.25 rem with $Q_a F=1.7$, as recommended in ICRP 10 and 10A reports.[12,14]

Probabilities of Intake

In attempting to derive the tritium activities handled, or in process, that would have an appreciable probability of resulting in the above intake (3 mCi T_2O or HTO per quarter), one is immediately confronted with a variety of parameters that affect the derivation. In many cases these parameters cannot be estimated satisfactorily. Moreover, the potential for a serious accidental exposure is very difficult to assess even when all the conditions are factored in and impossible

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*Immersion doses, with the skin as the critical organ, are not considered here.

in a case only vaguely or generally defined. For these reasons, some conservatism must be built into such estimates. Thus, conservative assumptions regarding credible accidents have been used to estimate activity levels in process, together with the experience of bioassay programs in routine operation in various installations, in order to select activity levels requiring bioassay. Some of the experience relating activity processed to intake is given in Table 1.

Levels in Process Requiring Bioassay

The levels of operation with tritiated water above which bioassay checks or routine evaluations have been carried out have varied considerably between institutions.(4) In some instances there is evidence that requirements have been too stringent and have resulted in unnecessary costs for more analysis than a safe surveillance program should have.(4) In other cases, the lack of a suitable bioassay program at an early date may have partly been responsible for serious overexposures.(5) In one reference, levels of operation above 10 curies of HTO in volatile or dispersible form have been suggested to require consideration of at least occasional bioassay whenever there was a possibility of release and inhalation of the material in process, in a volatile or readily-dispersible form; this 10 curie level was obtained partly by considering experience with plutonium and scaling up quantities eight orders of magnitude according to the relative dose per unit activity inhaled.(1) However, experience has shown that tritium as HTO can also penetrate certain protective gloves and be taken in through the skin unless gloves are washed frequently.(2) Also, incidents with smaller quantities of tritium have yielded measurable exposures that some institutions believe should be recorded for radiation monitoring purposes, even though these instances are rare and the cumulative exposures are small.

Experience from accidental releases does indicate that the probability of intake by inhalation would generally be much less than 10^{-5} of the material in process.[9,10] Assuming protection of hands and forearms in the case of tritium, these values from operational experience should also set a practical upper limit to the fraction of tritium inhaled compared to material in process. On the other hand, in many installations, since tritium as HTO can readily be taken up through skin or inhaled, and the intake of only about 5 millicuries could produce 1 rem (using a QF = 1.7), routine bioassay has often been provided at this level or below for open operations with little or no additional containment. There is considerable opinion that bioassay services at least on a sampling basis should be considered at open-bench levels of operation above 10 millicuries per worker in process, and should generally be required above 100 mCi in process, in order to achieve ALARA exposures, unless the nature of the operation insures additional containment or sufficient dilution with other process materials. Some experience with concentrations of tritium diluted in large volumes is also available from the author. Preliminary recommendations based on these values, and on minimum protection factors assumed for fume hoods and glove boxes, are presented in Table 2 for HTO. These values were selected to balance the apparent differences in practice and opinion, and provide a sound basis for establishing bioassay programs. Evidence reviewed in a supplementary document indicates that values a factor of 10 lower for tritiated nucleotide precursors, and values 1000 times higher for HT or T_2 gas in sealed containers preventing oxidation, than those in Table 2 would be appropriate for general guidance. Other compounds of tritium may be considered to be in approximately the same radiotoxicity category as HTO. Your comments on these suggested guidelines would be gratefully received.

Table 1

EXPERIENCE WITH INTAKE OF TRITIUM FROM VARIOUS PROCESSES

Type of Industry or Activity	Form of Tritium	Protective Enclosure	Significant Quantity Requiring Protection or Bioassay (Ci)	Activity in Process at One Time (Ci)	Activity Throughput Per Month (Ci)	Average Urine Levels of Workers	Estimated Fractional Intake of Activity in Process
LUMINOUS TIMEPIECE (Reference 17)	LUMINOUS PAINT, ORGANIC POLYMER	HOOD, BENCH COVERS	ALL PERSONNEL		10	14μCi/l	6×10^{-6} PER EMPLOYEE; 1.2×10^{-4} FOR 20 EMPLOYEES
LUMINOUS TIMEPIECE (USING CLOSED PENS) (Reference 18)	LUMINOUS PAINT, ORGANIC POLYMER	OPEN BENCH, MODIFIED WITH LOCAL EXHAUST	ALL PERSONNEL		10	2-3μCi/l	6×10^{-7} PER EMPLOYEE; 2×10^{-6} FOR THREE EMPLOYEES
LUMINOUS PAINT, MIXING POLYMER, PHOSPHOR, AND BINDER (Reference 19)	LUMINOUS PAINT, ORGANIC POLYMER	GLOVEBOX, SINGLE THICKNESS RUBBER GLOVES, SEVERAL HOURS OPERATION	ALL PERSONNEL	USUALLY 10 AT A TIME	800	10-20μCi/l	10^{-7} PER EMPLOYEE, MAINLY ONE EMPLOYEE
LUMINOUS PAINT, RESEARCH, DEVELOPMENT AND PRODUCTION (Reference 5)	GAS, HTO, VARIOUS ORGANICS	NONE--CARRIED OUT FOR SEVERAL YEARS			100-150	140-1,120μCi/l	4×10^{-6} TO 3×10^{-5} PER EMPLOYEE, TWO EMPLOYEES
	NOTE: Lethality produced in this case, presumably caused in part by tritium exposure.						
LUMINOUS WATCH STORAGE AREA - POOR VENTILATION (Reference 20)	SOLID PAINT ON TIMEPIECES	WATCH CRYSTALS, POORLY VENTILATED ROOM			17	2.7μCi/l (highest observed from storage and retailing)	7×10^{-7}

Continued on next page

Table 1 (continued)

EXPERIENCE WITH INTAKE OF TRITIUM FROM VARIOUS PROCESSES

Type of Industry or Activity	Form of Tritium	Protective Enclosure	Significant Quantity Requiring Protection or Bioassay (Ci)	Activity in Process at One Time (Ci)	Activity Throughput Per Month (Ci)	Average Urine Levels of Workers	Estimated Fractional Intake of Activity in Process
ACADEMIC LABORATORIES (Reference 15)	VARIOUS	USUALLY FUME HOOD	10mCi ON OPEN BENCH; 1Ci IN HOOD	MILLICURIES TO 1Ci	ABOUT 1Ci OR LESS	<1 uCi/l	$<5 \times 10^{-6}$ PER EMPLOYEE
ACADEMIC LABORATORY - SPILL (Reference 21)	HTO	SPILLED WITHIN HOOD: WIPES REMOVED FROM HOOD FOR DISPOSAL; GLOVES WORN (?)		0.078 - ALL SPILLED ON HOOD BENCH-TOP	SINGLE INCIDENT	14μCi/l PEAK; 100 MREM TOTAL DOSE	ABOUT 0.01 (HIGHEST OBSERVED FOR INCIDENT)
PARTICLE ACCELERATORS (References 18-20)	ADSORBED GAS AND HTO	DISASSEMBLED TARGETS AND ACCELERATOR PARTS	SURFACES EXPOSED TO 100Ci of GAS	100-200Ci		<3μCi/l ANNUALLY	$<2 \times 10^{-6}$ PER EMPLOYEE
TRITIUM GAS PROCESSING (Reference 25)	GAS (HT or T_2)	SPECIAL GLASS VESSELS AND GLOVE BOXES	ALL TRITIUM WORKERS	10^6Ci OR MORE	10^6Ci OR MORE	<28μCi/l	$<10^{-10}$
REACTOR OPERATIONS (References 26,28-31)	HTO DILUTED IN COOLANT	PRIMARY COOLANT SYSTEM EXCEPT FOR LEAKS	>0.01Ci/Kg FOR LWR'S	DEPENDS ON REACTOR DESIGN AND OPERATION		<1μCi/l (LWR'S)	
			>0.1Ci/Kg FOR HEAVY WATER REACTORS, WITH PLASTIC SUITS*	>10^5Ci FOR HEAVY WATER REACTORS		10μCi/l FROM 1Ci/Kg IN D_2O MODERATION (Reference 31)	10^{-5} OF CONCENTRATION IN COOLANT; 10^{-10} OF ACTIVITY IN REACTOR

*Urine levels would be 10-50 times greater without the plastic suits, which would make urine bioassay advisable at about the same concentration level (0.01Ci/Kg) as for LWR's without protection (Ref. 31).

Types of Operation	HTO Form or Compounds other than Nucleotides*	
	<10 Kg*	≧10 Kg
PROCESSES IN OPEN ROOM OR BENCH, WITH POSSIBLE ESCAPE OF TRITIUM FROM PROCESS VESSELS.	100 mCi	0.01 Ci/Kg
PROCESSES WITH POSSIBLE ESCAPE OF TRITIUM, CARRIED OUT WITHIN A FUME HOOD OF ADEQUATE DESIGN, FACE VELOCITY, AND PERFORMANCE RELIABILITY.	1 Ci	0.1 Ci/Kg
PROCESSES CARRIED OUT WITHIN GLOVEBOXES, ORDINARILY CLOSED, BUT WITH POSSIBLE RELEASE OF TRITIUM FROM PROCESS VESSELS AND OCCASIONAL EXPOSURE TO CONTAMINATED BOX AND BOX LEAKAGE.	10 Ci	1 Ci/Kg

*Nucleotide precursors - X0.1; HT or T_2 - X1000 in closed non-reactive systems.

Table 2

ACTIVITY LEVELS OR CONCENTRATIONS ABOVE WHICH BIOASSAY SHALL BE REQUIRED

INHALED FRACTIONS OF TOTAL ACTIVITY HANDLED (IFTAH)

	Glove boxes	Ventilated areas	Non-ventilated areas
SOLIDS AT AMBIENT TEMPERATURE	$1 - 10 \times 10^{-8}$ 2×10^{-8} *	$3 - 100 \times 10^{-8}$	$7 - 20 \times 10^{-7}$
AQUEOUS SOLUTIONS AT AMBIENT TEMPERATURE		$2 - 50 \times 10^{-8}$	
ELEVATED TEMPERATURES		$2 - 10 \times 10^{-6}$	

*IFTAH experimentally determined at Harwell.

From Franke and Hunzinger, 1968.

Table 3

QUESTIONNAIRE ON FACTORS DETERMINING REQUIREMENTS FOR H-3 BIOASSAY

Please mail to:

Dr. Allen Brodsky
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Telephone: (301) 443-6920

1. List any experience below that may be helpful in judging probabilities of intake. Attach frequency distributions of urine sample concentrations or interpreted dose where available. Include and identify accidental single intake as well as chronic exposures.

Form of tritium and type of process; protective facilities and equipment	Quantity in Process for each worker at any one time	Throughput in Curies per Year	Average Urine Concentration	Average or Single Intake Dose and/or IFTAH*

2. On the back of this sheet provide any estimates available on fractional amounts of activity in process that were taken into the body in accidental releases or spills. Give some details of the incident in each case.

3. Provide a brief description of your bioassay program and procedures, including criteria for including persons in the program and action levels. Attach any available written description of your program or reprints that are pertinent.

4. Provide any comments on levels requiring bioassay as proposed in this paper.

Thank you.

*IFTAH = Inhaled fraction of total activity handled (after Franke and Hunzinger, 1968)

EXHIBIT - NONE OF THESE INCIDENTS INVOLVED TRITIUM

Table 1. Inhaled Fraction of Total Activity Handled (IFTAH) for Accidents Involving some Types of Operation

	Glove boxes	Ventilated areas	Non-vent. areas
Solid compound Ambient temp.	1×10^{-7} (1)* 1×10^{-6} (2)	3×10^{-6} (3) 2×10^{-7} (4) 4×10^{-7} (5) 2×10^{-7} (15) 1×10^{-6} (19)	7×10^{-7} (7) 2×10^{-6} (20)
Aqueous sol. Ambient temp.		5×10^{-7} (6) 4×10^{-7} (8) 2×10^{-5} (9)	
Elevated temperatures		7×10^{-6} (10) 1×10^{-5} (11) 3×10^{-6} (12) 2×10^{-6} (13) 1×10^{-5} (16)	

* Numbers in brackets denote number of accident (see appendix).

explained by an example. Assume that the statistics yield a figure of $(1 \pm 0.5) \times 10^{-7}$ for the inhaled fraction in accidents during operations involving aqueous solutions at ambient temperature in a ventilated area. Assume further that such an operation is carried out with 10 mCi of ^{90}Sr, for which the dose equivalent to the critical organ has a value of 3.8×10^{7} rem/Ci. Multiplying these three figures, as explained above, yields a hazard for this operation of

$$H = (4 \pm 2) \times 10^{-2} \text{ rem}$$

This figure means that as a result of any accident during an operation in these conditions the operator's skeleton would probably receive a dose equivalent of 40 ± 20 mrem.

Similarly the opposite question of the maximum working limit of activity to be handled can be answered. Assuming the same type of operation and allowing for an accidental dose equivalent of 25 rem in the following 50 years, it follows that the maximum amount to be handled should be smaller than 6 Ci ^{90}Sr.

It is obvious that this type of analysis does not produce figures for the probability of an accident occurring, but it can predict the consequences in terms of dose equivalent. This information should not be neglected—if it is available.

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